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# Summary Report on Emissions from the Glass Manufacturing Industry

Battelle Columbus Labs, OH

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# Summary Report on Emissions from the Glass Manufacturing Industry

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This project was undertaken to evaluate emissions rates from typical glass manufacturing furnaces. The effort concentrated on the container segment of the industry, however, tests were also conducted on the pressed blown, and flat glass segments of the industry. The quantitative results of the test program were compared to earlier calculated results derived in the source assessment documents for each segment of the industry. Additional data collected during this test program included particle size distributions of glass furnace emissions and trace metals analyses of glass furnace emissions. Other sources within the typical glass manufacturing plant were also evaluated to determine the types of pollutants that are generated from these sources.

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# SUMMARY REPORT ON EMISSIONS FROM THE GLASS MANUFACTURING INDUSTRY

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## FOREWORD

When energy and material resources are extracted, processed, converted, and used, the related pollutional impacts on our environment and even on our health often require that new and increasingly more efficient pollution control methods be used. The Industrial Environmental Research Laboratory - Cincinnati (IERL-Ci) assists in developing and demonstrating new and improved methodologies that will meet these needs both efficiently and economically.

The purpose of this report is to present data and conclusions from a comprehensive testing program on glass melting furnaces. Previous EPA and industry reports on this subject have relied primarily upon literature and randomly collected data without regard to similitude of testing procedures and experimental design. Extreme caution, therefore, was taken on this program to ensure that each test was conducted in a similar manner and that each site tested reflected a typical operating situation in the industry. Wherever statistically possible, conclusions were developed to support or refute past conclusion on industry emissions.

The results of the report, therefore, will be extremely valuable to R&D programs concerned with emissions from the glass manufacturing industry.

For further information concerning this subject, contact the Industrial Pollution Control Division, Metals and Inorganic Chemicals Branch.

David G. Stephan
Director
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Cincinnati

## ABSTRACT

The objective of the research described in this report was to determine the character of emissions from glass manufacturing operations. To accomplish that objective, standardized testing procedures were conducted at several glass plants. These plants included the three industry segments (glass containers, SIC 3221; flat glass, SIC 3229; pressed and blown glass, SIC 3229) that produce glass articles through melting of raw materials and scrap glass. The products of purchased glass, (SIC 3231), and mineral wool, (SIC 3296), segments were specifically excluded because the former had no melting operations and the latter had melting operations vastly different from the other three. Also, textile fiber operations were specifically excluded.

This report presents a comprehensive review of the emission testing data. For the purposes of this report, the glass manufacturing process has been divided into five subprocesses: batching and mixing, melting, forming, post forming, and product packaging. This report presents new information about the size distribution of fine particulate matter emitted from the glass melting subprocess, especially with regard to the distribution of particulates considered to be respirable (i.e., $\leq$ 3 μm diameter). This report also presents important new information about the type of emissions issuing from some of the forming and post-forming process steps. Finally, the pertinent data from the field testing are compared to similar data that has been reported in the Source Assessment Documents for the three glass making segments.

This report was submitted in fulfillment of Contracts 68-01-3159 and 68-01-4431 by Battelle's Columbus Laboratories under the sponsorship of the U.S. Environmental Protection Agency. It covers the period from June 4, 1976 to September 1, 1978.

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# SECTION 1

# INTRODUCTION

## BACKGROUND AND OBJECTIVES

The Clean Air Act of 1977 requires the U.S. Environmental Protection Agency (EPA) to project the need to control emissions from a given industry. Source Assessment Documents (SAD's) are viewed as one means of making such projections. In the glass manufacturing industry, SAD's have been written for its three major Standard Industrial Classification segments--glass containers (SIC-3221),[1] flat glass (SIC-3211),[2] and pressed and blown glass, nec (SIC-3229).[3] These documents review the existing emissions information and are primarily based on data found in the National Environmental Data System (NEDS)[4] data bank.

This report evaluates information presented in the SAD's (Table 1) by using new data collected by onsite testing. The immediate object of this project is to measure the emissions from all glass manufacturing operations in all three major segments of the industry within the limits of available time and funding. Emission measurements have been taken in thirteen stacks at seven plants.

A number of problems were encountered during the course of the study. Chief among them was the fuel crisis brought about by the severe winter of 1975-76. Many companies that had indicated willingness to participate were unable to do so because of fuel curtailments. Others were prevented from participation by plant shutdown or manufacturing problems that could not be solved within the time available.

## TEST METHODS

For the purposes of this research program, the glass manufacturing operation was divided into five processes: batching and mixing, melting, forming, postforming, and product packaging. Because of the low levels of emissions for batching and mixing and product packaging, these process steps were tested. Standard, EPA-recommended testing procedures were used to establish the emission rates of criteria pollutants from the melting process, and a porous-polymer adsorbent trap has been used for Level-I testing for organic emissions from the forming and postforming process steps.

TABLE 1. SUMMARY OF EMISSION INFORMATION AVAILABLE IN SOURCE ASSESSMENT DOCUMENTS

| Type Of Emission | No. Of Cases | Percent of Total Furnaces |
|---|---|---|
| Glass containers (325 furnaces): | | |
| Particulates: | | |
| Loading | 89 | 27 |
| $SO_x$ | 49 | 15 |
| $NO_x$ | 24 | 7 |
| HC | 36 | 11 |
| Pressed and blown glass (383 furnaces): | | |
| Particulates: | | |
| Loading | 77 | 20 |
| $SO_x$ | 59 | 15 |
| $NO_x$ | 63 | 16 |
| CO | 55 | 14 |
| HC | 55 | 14 |
| Flat glass (29 furnaces): | | |
| Particulates: | | |
| Loading | 5 | 17 |
| $SO_x$ | 3 | 10 |
| $NO_x$ | 3 | 10 |

## SECTION 2

## CONCLUSIONS AND RECOMMENDATIONS

Some of the test data from this research program disagrees with those of the SAD's. The magnitude of the disagreement varies for each criteria pollutant, and no particular trends are evident for either pollutants or industry segments. (See Section 5)

Several interesting results have been established for the melting emissions, over a range of compositions including soda-lime, borosilicate, and lead glass. They also span process rates from 811 to 15,900 kg/hour. Consequently, these results are expected to be generally applicable to all three industry segments. First, statistically significant, empirical (regression) relationships have been established for (1) $SO_x$ emission rates as a function of process rate and fuel use rate, (2) for $NO_x$ emission rates as a function of fuel use rates, and (3) for total particulate emission rates as a function of fuel use rates. For the tested plants, these relationships are not strongly influenced by furnace size, type of glass melted, or type of fuel used. Also, more than 80 percent of the total particulate matter is respirable (i.e., median diameter $\leq 3\mu m$), and approximately 15 percent of that total particulate matter is formed by condensation or reaction with the impingers in the EPA-5 sampling train. Finally, emissions of unburned hydrocarbons and carbon monoxide are negligible.

The Level-I testing for organic emissions has revealed that 47 percent of the emissions from the forming and postforming process steps are composed of the following species:

- Aliphatic hydrocarbons
- Aromatic hydrocarbons
- Polyorganic matter (POM)
- Polychlorinated biphenyls (PCB)
- Esters
- Ethers
- Nitrocompounds
- Halides

An additional 39 percent of the emissions may be composed of one or all of the following:

- Sulfonates
- Sulfoxides
- Sulfonic acids.

The emissions from the latter list may be preferentially produced from the postforming process. However, caution must be exercised in interpreting these results because they are not definitive. They indicate only a potential problem area and not the severity of the potential problem.

A comparison of forming and postforming organic emissions measured in this test program and the values reported in the SAD's is inappropriate because a Level-I test does not define emission rate. Also, the data in the SAD's are reported as engineering estimates from mass balance calculations and may not reflect actual conditions. Therefore, any comparison of data from these two sources can be erroneous. The Level-I testing has revealed a list of toxic substances that may potentially be emitted during the manufacture of glass articles. The rate, severity, and exact character of these emissions has not been established. More testing is therefore required before the need for forming and postforming emission control can be established. If control of these emissions is required, then the only economically and technically feasible solution is replacement of the oils and sprays that generate these emissions. Such a replacement will require a considerable technical development effort.

Other sources of emission from the forming and postforming process steps are acid fumes and metal (particularly tin) vapors. Actual testing for these species was not conducted during this program because permission was unobtainable for testing sites that could potentially emit those species. However, data that have been supplied by the participating manufacturers indicate that the information reported in the SAD's adequately estimates these emissions.

The control technology available for the melting operation seems to control particulate emissions adequately within current regulations. But, these devices may not be adequate as the regulations become more stringent. The fine particulate emissions are especially important when reviewed with general information about the efficiency of control devices as follows:

GENERAL EFFICIENCIES OF CONTROL DEVICES FOR PARTICULATE MATTER [5]

| Device | Efficiency by Median Particle Size | | |
|---|---|---|---|
| | 0.5 µm | 1 µm | 5 µm |
| Baghouse | 90 percent | 95 percent | > 95 percent |
| Venturi Scrubber | 80 percent | 95 percent | 95 percent |
| ESP | <70 percent | 75 percent | 90 percent |

The electrostatic precipitator (ESP) is emerging as the preferred control device in the industry for several technical reasons. However, it may be ill-suited for control of the fine particulate matter issuing from glass melting furnaces. The $NO_x$ and $SO_x$ emissions present difficult control problems because proven control devices for these species are unavailable. Electric melting may offer a means of reducing these emissions, but that technology has been demonstrated only for smaller melting units (100 to 110 Mg/day) and a development effort would be required to raise the capacity of electric melting to the nominal rate of 220 Mg/day. Implementation of electric melting in the flat glass industry poses an even larger problem because the technology must be scaled to units that produce glass at a rate of 600 to 800 Mg/day. Other technologies such as batch preheating and oxygen enrichment may offer potential solutions to these problems, but they require considerable development and evaluation before they can become commercially available. Reasonable control over HC and CO emissions can be accomplished by adequate combustion control. Equipment and combustion monitoring devices are commercially available for accomplishing that control.

## SECTION 3

## INDUSTRY DESCRIPTION

The glass industry consists of five segments: flat glass, glass containers, pressed and blown glass, mineral wool and products of purchased glass (SIC's 3211, 3221, 3220, 3296, 3231). Table 2 gives the value of shipments, employees, and plant numbers estimated for 1971 and 1973 for four of these segments. In 1973, some 1,335 plants employing more than 200,000 persons produced merchandise valued at $6.3 million. The glass container segment is the largest in terms of shipments and employees. Approximately 57 percent of all glass melted is produced by the glass container segment. The remaining glass melting is roughly divided between the flat glass (24 percent) and pressed and blown glass (19 percent) segments. The products of purchased glass segment has no melting operations (the primary emissions source in glass manufacturing), thus that industry has not been included in this study. The melting process for the mineral wool industry is a cupola process which is vastly different from the traditional, fossil fuel glass melter, consequently, this industry segment has been excluded from this study. Textile fibers are included in SIC-3229, but the contract for this project specifically excluded that portion of SIC-3229 from testing.

TABLE 2. ESTIMATED SHIPMENTS, EMPLOYEES, AND PLANT NUMBERS FOR THE GLASS INDUSTRY, 1971 AND 1973

| SIC No. | Industry segment | Value of Shipments (Millions of $) 1971 | 1973 | Change (%) | Employees (Thousands) 1971 | 1973 | Change (%) | Number of plants |
|---|---|---|---|---|---|---|---|---|
| 3211 | Flat glass | 811 | 1,118 | 37.8 | 24.0 | 26.3 | 9.6 | 75 |
| 3221 | Glass containers | 1,944 | 2,316 | 19.1 | 74.6 | 77.8 | 4.3 | 140 |
| 3229 | Pressed and blown glass | 1,108 | 1,423 | 28.4 | 53.4 | 61.9 | 15.9 | 260 |
| 3231 | Products of purchased glass | 1,157 | 1,440 | 24.4 | 29.8 | 35.5 | 19.1 | 860 |
| | Total | 5,020 | 6,297 | | 181.8 | 201.5 | --- | 1,335 |
| | Average | --- | --- | 25.4 | --- | --- | 10.8 | --- |

(a) Source: Department of Commerce

## GLASS CONTAINERS (SIC-3221)

Glass containers (SIC-3221) is the largest of the three major segments of the glass industry. It includes the manufacture of narrow-neck and wide-mouth glass containers for foods, beverages, medicines, toiletries, and cosmetics. Shipments in this segment have grown at an average rate of about 3.5 percent since 1971. Three general types of container glass are produced: amber, green, and clear. For this report, green and clear glass are considered together in a single category designated "flint". The major difference between amber and flint is the iron-oxide additions. Amber accounted for approximately 15 percent of glass production in 1976 and flint represented about 85 percent.

According to information gathered from the Department of Commerce 1972 Census of Manufacturers and from the Glass Packaging Institute (GPI), 119 establishments manufacture glass containers in the United States. These 119 plants are operated by 27 manufacturers (Table 3). Statistics obtained from 1976 glass industry directories and from industry sources indicate that approximately half of these plants are operated by the five largest companies. Geographically, glass container plants are located near the local markets they serve. Plants are found throughout the United States, but a large number are concentrated in the East, North-Central, and Middle Atlantic regions. The regional distribution of the major plants is shown in Figure 1.

The volume of shipments for 1976 was 293 million gross compared to 268 million gross in 1972, a 9-percent increase for the 4 year period. The weight of glass containers shipped increased from 10,772 Gg in 1972 to approximately 11,809 Gg in 1976, an increase of 10 percent.

The basic raw materials for soda/lime container glass are silica sand, soda ash ($Na_3CO_3$), and limestone (primarily $CaCO_3$, plus some $MgCO_3$ in dolomitic limestones). Feldspathic minerals (anhydrous aluminosilicates containing potassium, sodium, and calcium in varying ratios) are also utilized as sources of alumina and alkali. Minor amounts of other oxides are introduced as impurities, and additional minor ingredients are added for specific purposes. A typical soda-lime glass-batch composition is:

| | |
|---|---|
| Silica sand | 909 |
| Soda ash | 306 |
| Feldspar | 118 |
| Limestone | 294 |
| Salt cake ($Na_2SO_4$) | 7 |
| Total | 1,634 kg |

TABLE 3. MAJOR GLASS CONTAINER MANUFACTURERS IN THE UNITED STATES[a]

| Manufacturer | No. Of Plants | Manufacturer | No. Of Plants |
|---|---|---|---|
| Anchor Hocking | 9 | Kerr Glass | 7 |
| Arkansas Glass | 1 | Latchford Glass | 1 |
| Ball Corporation | 4 | Leone Industries | 2 |
| Bartlett Collins | 1 | Liberty Glass | 1 |
| Brockway Glass | 14 | Madera Glass Co. | 1 |
| Chattanooga Glass | 7 | Metro Glass | 4 |
| Columbia Gas | 1 | Midland Glass | 4 |
| Diamond Glass | 1 | National Bottle Corp. | 4 |
| Gallo Glass | 1 | National Can | 4 |
| Glass Containers Corp. | 12 | Owens-Illinois | 20 |
| Glenshaw Glass | 2 | Thatcher Glass | 6 |
| Hillsboro Glass | 1 | Underwood Glass | 1 |
| Indian Head | 7 | Wheaton Glass | 2 |
| Industrial Glass | 1 | Total | 119 |

a) Source: Material provided from GPI, dated October 24, 1975.

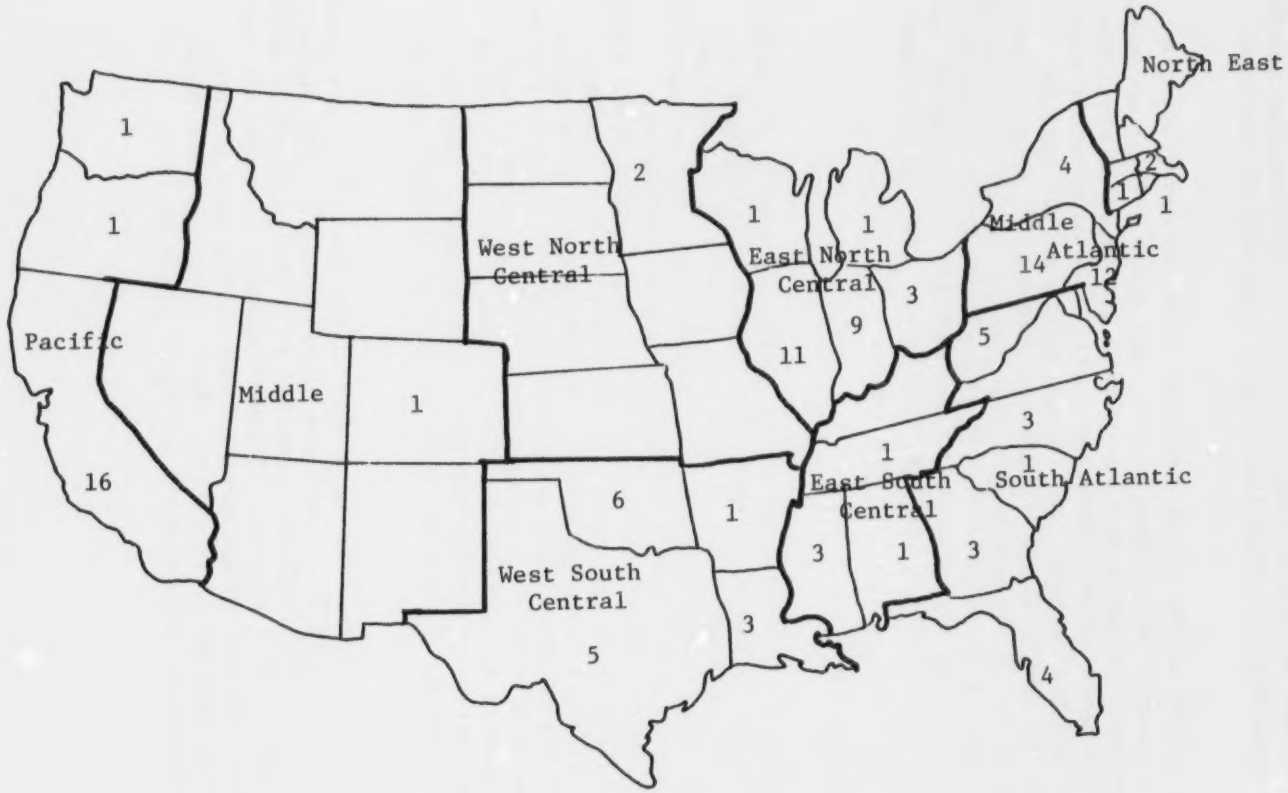



Figure 1. Regional distribution of glass container manufacturers.

Typically, these ingredients will produce 1,370 kg of glass and give off 259 kg of gases, primarily (>99 percent) $CO_2$. The batch volume of 1.2 $m^3$ will produce 0.6 $m^3$ of fluid glass and 858 $m^3$ of gaseous products (measured at the furnace temperature of 1,500 C). The minor ingredients such as salt cake (sodium sulfate) and various fining, coloring, or decolorizing agents, rarely exceed 5 percent and are often less than 0.1 percent of the total glass composition.

Historically, growth of the glass container industry has fluctuated considerably, but shipments have steadily increased since 1967 at an average annual rate of 6 percent. A large portion of this growth is attributable to the popularity of the nonreturnable beverage bottle. In more recent years, growth in shipments has been slower, and future growth may be tied to legislation restricting use of nonreturnable containers. Production levels for 1980 are likely to be 20 percent higher than levels for 1974. Emissions are expected to increase proportionally, and possibly at an increased rate, since the industry is moving from the use of natural gas to the use of fuel oil. The actual effect of this conversion on emission rates is not clear.

## PRESSED AND BLOWN GLASS, NEC (SIC-3229)

The pressed and blown glassware industry, as represented by SIC-3229, essentially includes all industrial establishments primarily engaged in manufacturing glass and glassware that is pressed, blown, or shaped from glass produced in the same establishment. It consists of every category of glass or glassware except flat glass (SIC-3211) and glass containers (SIC-3221). Establishments include those manufacturing textile glass fibers; lighting, electronic, and technical ware; and machine-made and handmade table, kitchen, and art-ware glass products. The pressed and blown glass industry consists of approximately 155 manufacturing establishments (Table 4). Approximately 40 plants produce hand pressed and blown glassware almost exclusively. The industry is concentrated in or about the North Central region of the United States--primarily New York, Pennsylvania, West Virginia, Ohio, Indiana, and Illinois. Plants are also located in 22 other states (Figure 2).

Each of three product types makes up a significant portion of the pressed and blown glass industry (Table 5). The value of shipments for the industry in 1976 is estimated to be $1.6 billion, compared with $1.1 billion in 1971--an average annual increase of 9 percent. Shipment weights from the various industry segments are difficult to estimate because of the many product types and the different methods of reporting. But, the pressed and blown glass segment is estimated to have shipped 1.5 Tg of glass in 1976.

TABLE 4. MAJOR PRESSED AND BLOWN GLASSWARE MANUFACTURERS IN THE UNITED STATES

| Manufacturer | No. of Plants | Manufacturer | No. of Plants |
|---|---|---|---|
| American Optical | 3 | Kessler, Inc. | 1 |
| Anchor Hocking | 4 | King Seeley Thermos Company | 2 |
| Arrowhead Puritas | 1 | Labino Glass Laboratories | 1 |
| Atlantic Optical Moulding | 1 | Lancaster Glass Corporation | 1 |
| B and J Optical | 1 | Lenox Crystal | 1 |
| Bartlett-Collins | 1 | Lewis County Glass | 1 |
| Bausch and Lomb | 1 | Louie Glass Company | 1 |
| Beaumont Company | 1 | Marble King | 1 |
| Big Pin Glass Company | 1 | Masden Ind. | 1 |
| Blenko Glass Company | 1 | Mayflower Glass | 1 |
| Brock Glass Company | 1 | Mid-Atlantic | 1 |
| Brockway Glass | 2 | J. H. Millstein | 1 |
| E. O. Brody Company | 1 | Minners | 1 |
| Cambridge Glass | 1 | Multicolor Glass | 1 |
| Canton Glass | 1 | Nuclear-Pacific | 1 |
| Cataphote | 1 | Overmyer-Perram | 1 |
| Champion Agate Company | 1 | Owens-Illinois | 10 |
| Chemglass, Inc. | 1 | Pacific Glass Works | 1 |
| Corning Glass Works | 14 | Peltier Glass Company | 1 |
| Crescent Glass Company | 1 | Penbarthy Electromelt Intl., Inc. | 1 |
| Crystal Art Glass | 1 | Pennsboro Glass Company | 1 |
| Davis-Lynch Glass Company | 1 | Phoenix Glass Company | 1 |
| EMC Glass Corporation | 1 | Pikes Peak Glass Company | 1 |
| Eastman Kodak Company | 1 | Pilgrim Glass Corp. | 1 |
| Emerson and Cuming | 1 | Pittsburgh Corning Corp. | 2 |
| Elite Company, Inc. | 1 | Pape Scientific, Inc. | 1 |
| Erie Glass Mfg. Company | 1 | Potter Industry, Inc. | 4 |
| Erskine Glass and Manufacturing | 1 | RCA Corp. | 1 |
| Federal Glass Company | 1 | Rainbow Art Glass | 1 |
| Fenton Art Glass | 1 | Ray-Lite Glass, Inc. | 1 |
| Fischer and Porter Company | 2 | Reha Glass Company | 1 |
| Fostoria Art Glass | 1 | Rocky Mountain | 1 |
| Friedrich and Dimmock, Inc. | 1 | Rodifer Gleason Glass Corp. | 1 |
| GTE Sylvania, Inc. | 4 | St. Clair Glass Works | 1 |
| General Electric | 9 | Scandia Glass Works | 1 |
| Gentile Glass Company | 1 | Schott Optical Glass Company | 1 |
| Gillinder Brothers | 1 | Scott Depot Glass Company | 1 |
| Gladding-Vitro-Agate Company | 1 | Sellstrom Mfg. | 1 |
| Glass Works, Inc. | 1 | Earl Shelly Glass Company | 1 |
| Guernsey Glass Company | 1 | Sinclair Glass | 1 |
| Haley Glass Company | 1 | L. E. Smith Glass Company | 1 |
| Hamon Handcrafted Glass | 1 | Super Glass Corp, | 1 |
| Harvey Ind. | 1 | Swift Glass Corp. | 1 |
| Holophane | 1 | Techniglass, Inc. | 1 |
| Houze Glass Corp. | 1 | Thermal American Fused Quartz | 1 |
| Imperial Glass Corp. | 1 | Thomas Ind., Inc. | 1 |
| Indiana Glass Corp. | 1 | Variety Glass, Inc. | 1 |
| Industrial Glass and Plastics | 1 | Venejian Art Glass | 1 |
| Interpace Corp. | 1 | Victory Glass Company | 1 |
| Jeanette Corporation | 1 | Viking Glass Company | 1 |
| Jeanette Shade and Novelty Co. | 1 | West Virginia Glass Specialty Co. | 1 |
| Johnson Glass | 1 | Westinghouse Electric Corp. | 1 |
| Kanawa Glass Company | 1 | Westmoreland Glass Company | 1 |
| Kapp Glass, Inc. | 1 | Wheaten, Ind. | 1 |
| Kaufman Glass Company | 1 | The Paul Wissmach Co., Inc. | 1 |




Figure 2. Regional distribution of pressed and blown glassware manufacturers.

TABLE 5. PROPORTION OF INDUSTRY OUTPUT ACCOUNTED FOR BY THE CONSUMER, SCIENTIFIC, TECHNICAL, AND INDUSTRIAL GLASSWARE SEGMENTS OF SIC-3229[a]

| Process and Major Products | SIC | % of Industry Shipment (value) 1972 | 1976 |
|---|---|---|---|
| Table, Kitchen, and Art Ware | | | |
| Machine Made | (32291) | 33 | 39 |
| Hand Made | | 8 | 6 |
| Lighting and Electronic | (32292) | 35 | 32 |
| Scientific and Industrial | (32294) | 24 | 23 |
| Total Industry (Percent) | | 100 | 100 |
| (Actual) | | $1.1 billion | $1.6 billion |

(a) Source: Current Industrial Reports, MA-32E for shipment value in SIC-32291, -32292, and -32294 -- 1972 and 1976.

Compositionally, the four important categories of glass manufactured by the pressed and blown glass industry are as follows:

| Glass category | Estimated percent of total production |
|---|---|
| Soda/lime | 77 |
| Borosilicate | 11 |
| Lead silicate | 5 |
| Opal | 7 |

Soda/lime glasses are overwhelmingly the most important type of glass in terms of variety of use as well as tonnage melted. The combination of silica sand, soda ash, and limestone produces a glass that is easily melted and shaped and that has good chemical durability. In the soda-lime-silica system, an optimum glass with respect to cost, durability, and ease of manufacture is typically composed as shown in Table 6. Magnesia is used primarily to reduce cost by the substitution of dolomitic limestone for limestone as a raw material. The alumina improves chemical durability and decreases the problem of crystallization during forming. Primary pressed and blown products employing this type of glass are incandescent lamps, tubing, and tableware.

Borosilicate glasses are basically a combination of silica sand, boric oxide, and soda ash. The compositional ranges of typical commercial borosilicates are shown in Table 6. The borosilicate glasses have excellent chemical durability and electrical properties, and their low thermal expansion yields a product with high resistance to thermal shock. These combined properties make them ideal for demanding industrial and domestic uses such as chemical laboratory ware, cookware, pharmaceutical ware, and some lens reflectors and lamp envelopes. Pyrex$^{R}$, produced by Corning Glass Works, and Kimax$^{R}$, produced by the Kimble Division of Owens-Illinois, Inc., are examples of products made from borosilicate glasses.

Lead silicate glasses are composed of silica, lead oxide, and significant amounts of alkali oxide. The compositional range of typical commercial lead glasses is shown in Table 6. The lead glasses are characterized by high electrical resistivity, high refractive index, and slow rate of increase in viscosity with decreasing temperature. This viscosity characteristic makes them particularly well suited to hand fabrication. Lead glasses are used in high-quality art and tableware, for special electrical applications, optical glasses, fluorescent lamp envelopes, and X-ray, gamma-ray, and neutron radiation shielding windows.

Opal glasses are translucent and may be colored. Commercial products of opal glass include lighting globes, ointment jars, dinnerware, and wall paneling. The translucency or opacity of opal glasses is produced by multiple scattering of light inside the glass. This scattering is achieved by the precipitation of crystals (or an immiscible amorphous phase) with an index of refraction different from that of the base glass. Commercial opal glasses commonly employ fluorine additions to yield opacifying crystals of sodium or calcium fluoride. A typical commercial opal glass jar composition is given in Table 6 along with an opal illumination glass composition.

TABLE 6. TYPICAL GLASS COMPOSITIONS

| Major Components | Percent By Weight |
|---|---|
| **Soda/lime glasses:** | |
| Silica | 72 |
| Soda | 15 |
| Lime and Magnesia | 10 |
| Alumina | 2 |
| Other Oxides | 1 |
| **Borosilicate glasses:** | |
| Silica | 60-81 |
| Alumina | 1-17 |
| Boron Oxide | 5-24 |
| Sodium Oxide | 1-15 |
| Magnesia plus calcia | 4-17 |
| Potassium Oxide | 1- 8 |
| **Lead glasses:** | |
| Silica | 35-70 |
| Lead Oxide | 12-60 |
| Sodium Oxide | 4-8 |
| Potassium Oxide | 5-10 |
| Alumina | 0.5-2.0 |

| Major Components | Percent By Weight |
|---|---|
| **Opal glass:** | |
| $SiO_2$ | 71.2 |
| $Al_2O_3$ | 7.3 |
| CaO | 4.8 |
| $Na_2O$ | 12.2 |
| $K_2O$ | 2.0 |
| $F_2$ | 4.2 |
| **Opal Illumination glass:** | |
| $SiO_2$ | 59.0 |
| $Al_2O_3$ | 8.9 |
| CaO | 4.6 |
| MgO | 2.0 |
| $Na_2O$ | 7.5 |
| $F_2$ | 5.0 |
| ZnO | 12.0 |
| PbO | 3.0 |

The pressed and blown glass industry is very diverse, and estimates of future growth trends are difficult to make. The diversity of the industry tends to exempt it from fluctuations caused by changes in other glass industry segments. An annual growth rate of 5 to 8 percent is not an unreasonable expectation for this segment.

## FLAT GLASS (SIC-3211)

This industry contains establishments primarily engaged in manufacturing flat glass as well as some laminated and tempered glass. The major products shipped by the flat glass industry are: window glass, plate and float glass, rolled and wire glass, tempered glass, and laminated glass. Tempered and laminated glass production is also reported under glass products made of purchased glass (SIC 3231).

Flat glass shipments vacillated significantly in the latter 1960's and early 1970's. The value of shipments during 1972 was $937.2 million--up 15.5 percent from the previous year. The large fluctuations were due to domination of the industry demand by the construction and automotive industries. The four largest companies in the flat-glass industry accounted for more then 92 percent of the total value of industry shipments. In 1973, the average value of shipments from each of the 31 establishments was approximately $1.4 billion. Two areas in the U.S. accounted for slightly more than 70 percent of the value of shipments made by all establishments in the industry--the North Central (42 percent) and the South Central (29 percent) regions. Most flat-glass manufacturing plants are located in Pennsylvania, Ohio, Tennessee, Michigan, Illinois, Texas, and California (Figure 3). The seven flat glass manufacturers are shown in Table 7, as well as the approximate market share (by product type) for each of these companies.

Four flat glass products--float, sheet, rolled, and plate--are manufactured in the United States. Of these float glass accounts for more than 90 percent of the total flat glass production. Float glass is made by floating molten glass from the melting furnace on a bath of molten tin until the glass hardens. This glass, with its high optical quality, has replaced plate glass, which required grinding and polishing to produce a smooth surface. It is used for automobile windows and large picture windows. Average thickness ranges from 3.2 to 6.4 mm. Sheet glass is made by drawing molten glass upward from the melt. It is thinner than float glass (1.6 to 3.2 mm) and is used for windows in residential construction. Rolled or patterned glass is formed by drawing molten glass through rollers with patterns impressed on them. This decorative glass is used for special purposes such as shower doors and partitions. Plate glass is made by drawing molten glass through smooth rollers and then grinding and polishing both glass surfaces to a smooth finish. Only one plate glass furnace is still in operation in the United States.

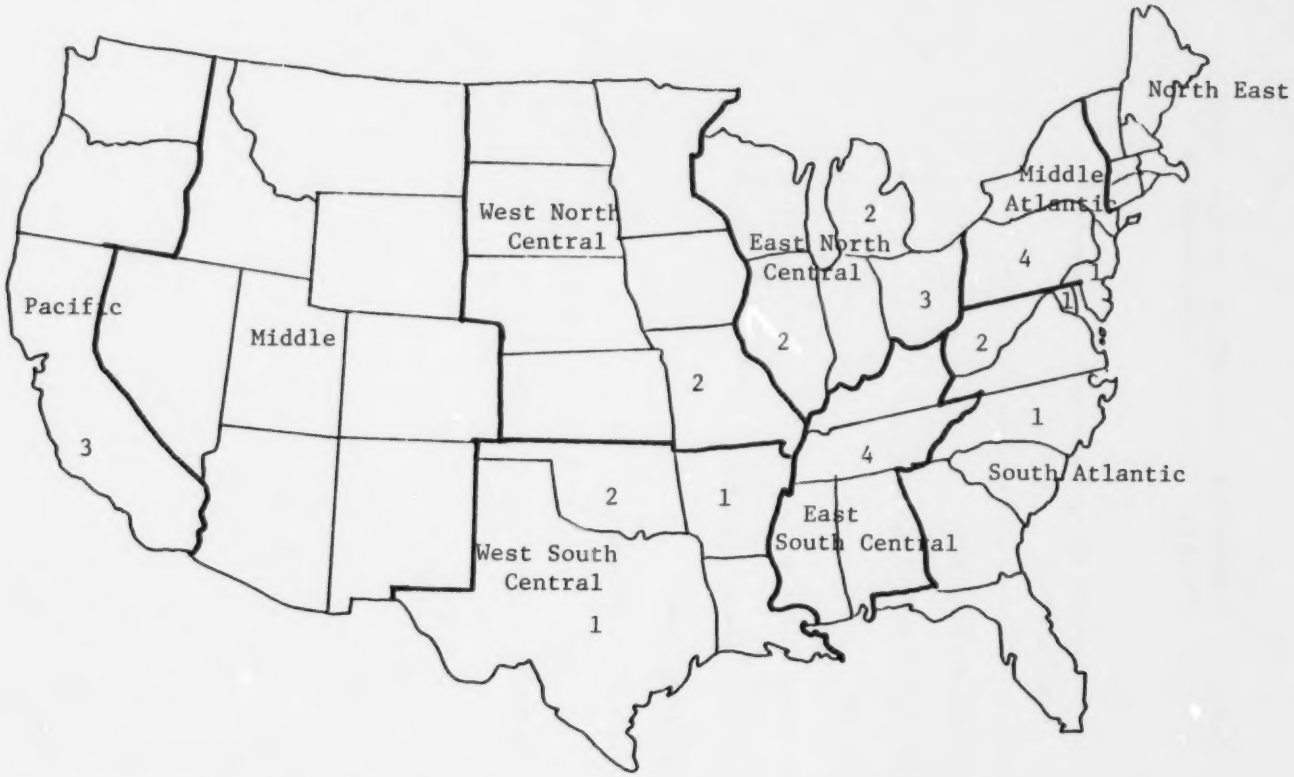



Figure 3. Regional distribution of flat-glass manufacturers.

TABLE 7. PERCENTAGE DISTRIBUTION OF FLAT GLASS CAPACITY BY SEVEN MAJOR COMPANIES(a)

| Manufacturer | |
|---|---|
| PPG Industries, Inc. | 45.1 |
| Libbey-Owens-Ford Company | 26.4 |
| Ford Motor Company | 13.1 |
| ASG Industries, Inc. | 6.3 |
| Guardian Industries Corp. | 3.0 |
| C-E Glass, Inc. | 1.5 |
| Fourco Glass Company | 4.6 |
| | 100.0 |

The composition of flat glass is exclusively soda-lime-silica with the following typical recipe:

| | kg | wt% |
|---|---|---|
| Silica sand | 910 | 56 |
| Soda ash | 302 | 18 |
| Feldspar | 80 | 5 |
| Limestone | 309 | 19 |
| Salt cake ($Na_2SO_4$) | 36 | 2 |
| Total | 1,637 | 100 |

These ingredients melt to 1,341 kg of glass and give off 295 kg of gases, primarily (>90 percent) $CO_2$. The batch volume of 1.27 $m^3$ produces 0.57 $m^3$ of fluid glass and 708 $m^3$ of gaseous products (measured at the furnace temperature of 1,500 C). Although many minor ingredients (<5 percent of the batch) can be added to the glass batch, very few are used in making flat glass. The only other ingredients used in making clear float glass are carbon in the form of powdered coal (used as a reducing agent for sulfates) and iron oxide (to provide a greenish tint). No borates, fluorides, selenium, or arsenic compounds are added. A small amount (<10 percent of total production) of colored flat glass is made, but that production has not been considered in this document.

The flat glass industry has sustained a compound annual growth for the value of shipments of 6.7 percent between 1967 and 1976. A continued growth rate averaging 5 percent is not unexpected. However, this industry is strongly tied to the housing and automotive industries and the future growth trends of those industries will greatly affect the flat glass industry.

## SECTION 4

## TEST METHODS

For the purpose of this project, the glass manufacturing operation has been divided into the following processes:

- Batching and mixing
- Melting
- Forming
- Postforming
- Product packaging

These processes generally hold for both the glass container and pressed and blown glass segments. In the flat glass industry, forming and postforming are usually integrated into one continuous process. The primary function of the postforming operation is to remove detrimental, residual stresses in the glass product, and in that sense, forming and postforming operations in the flat-glass manufacturing industry can be considered as being equivalent to similar operations in the other two glass manufacturing segments.

The test procedures that have been used in this program have generally been standard EPA procedures. Since this program is not a compliance testing program, some of the tests have been modified to maximize the information while minimizing the cost. The test procedures and modifications will be described as they apply to each process.

### BATCHING AND MIXING

The first process in the manufacture of glass articles is the weighing and mixing of the raw materials--sand, alkali, alkaline-earth carbonates, various mineral products, and cullet (scrap glass). Minor ingredient additions to the glass recipe include carbon and sulfates that provide a means for removing gaseous inclusions (refining), and various agents that impart color to the final product. The raw materials and cullet are accurately weighed, as specified by the particular glass recipe, and intimately mixed before delivery to the melting unit.

The major emission encountered in this process is dust that results from the transfer, handling, and weighing of the generally fine-grained raw materials. These emissions are either controlled through the use of a bag-house (in which case the control efficiency is >90 percent), or they are uncontrolled and become fugitive emissions. Batch wetting is sometimes used to reduce dusting during material transfer. If the control device is in

operation, there is no point in measuring the very low level of emission from that device. If on the other hand the dust is fugitive, there is no reliable means of measuring that emission. Therefore, no test data have been collected for this process.

## MELTING

The glass melting operation is a high-temperature combustion process and is the major source of emissions in the glass manufacturing operation. The emissions from this process include particulates, $NO_x$, $SO_x$, CO, and hydrocarbons. Both natural gas and fuel oil are used as primary fuels in this step. Where possible, the sampling program includes testing for both fuels. The test procedures for melting are listed in Table 8. In some cases, part of the raw material feed includes a fluoride flux. When a fluoride flux is used, the EPA-5 train is modified to include the impingers for an EPA-13 test. This modification allows an analysis of particulate fluorides. The particle-size distribution is determined by using an Anderson cascade impactor, which takes data at the stack temperature. This impactor is run at an average flow point, which is determined during a traverse of the stack for the particulate loading test. This average flow point is also used for the testing of $NO_x$, $SO_x$, and CO, and hydrocarbons. The integrated bag sample is also taken from this average flow point, and that bag is used to perform an Orsat analysis.

## FORMING

Both pressed and blown glass and glass containers are formed in automated machines. Various organic sprays and swabbing compounds are used in this process to facilitate release of the formed product from the hot metal mold. Volatilization of these compounds provides a source of emissions which exit from the area through ventilators located in the roof of the building. A Level-I test for the organic emissions is performed for this process step using a porous polymer adsorbent trap[6] (Figure 4). This test allows calculation of emissions concentration and determination of eight classes of organics (Table 9).

The forming process that is used in the manufacture of flat glass is considerably different from that used in pressed and blown glass and in glass containers. In flat glass manufacturing, the molten glass floats onto a bath of molten tin and is drawn into a very large flat sheet. The emissions from this process step are expected to be very minimal and are calculated on a worst-case basis using actual, annual use rates of tin.

## POSTFORMING

The postforming process includes annealing to remove detrimental, residual stresses in the glass products, lubricity coatings, and decorating. The lubricity coatings and decorating media are potential sources of organic emissions during postforming. These potential emissions are determined using

TABLE 8. SOURCE EMISSION TEST PROCEDURES FOR GLASS MELTING FURNACES

| No. of samples per source (runs) | Sample type | Sample method | Minimum sample time (min) | Minimum sample volume (scf) | Initial analysis | Final analysis |
|---|---|---|---|---|---|---|
| 3 | Particulates | EPA-5 [a] EPA-13[e] | 60 | 30 | Mass[b] 1/2 Mass[b] 1/2 Mass | Trace metals, OES-AA Trace metals, OES-AA Fluorides |
| 3 | Particle size | Anderson at one traverse point | ≈10[d] | ≈7[d] | Mass size | --- |
| 3 | Gases | EPA-10[c] at one traverse point | 60 | 1 | Orsat[f] | - |
| | | " | ≈15 sec | 2-liter flasks | GC[g] | - |
| | | " | ≈15 sec | 2-liter flasks | $NO_x$[h] | - |
| 1 | Gas | Grab | ≈15 sec | 2-liter flasks | GC[g] | - |
| 1 | Gas | EPA-7 | ≈15 sec | 2-liter flasks | $NO_x$[h] | - |
| 3 | $SO_x$&$SO_2$ | EPA-8 at one traverse point | 20 | 10 | $SO_x$&$SO_2$ | - |

a Twelve-point samples at 5 min/point, EPA-5 as in 6/8/76 Federal Register.
b Front and back half mass as per EPA-5 in 8/17/71 Federal Register.
c When flourides are present, use Method 5 train with EPA-13B solutions and fluoride analyses. One run for mass and 2 runs for fluoride.
d Approximate. Depends on stack gas particulate loading.
e Tedlar bag and flasks are black or covered and protected from light.
f Orsat or Fyrite; if CO is present, Orsat is required.
g GC analysis through C-4 (one flask analyzed per source other backup).
h EPA-7 flask for phenoldisulfonic acid method (one flask analyzed per source other backup).

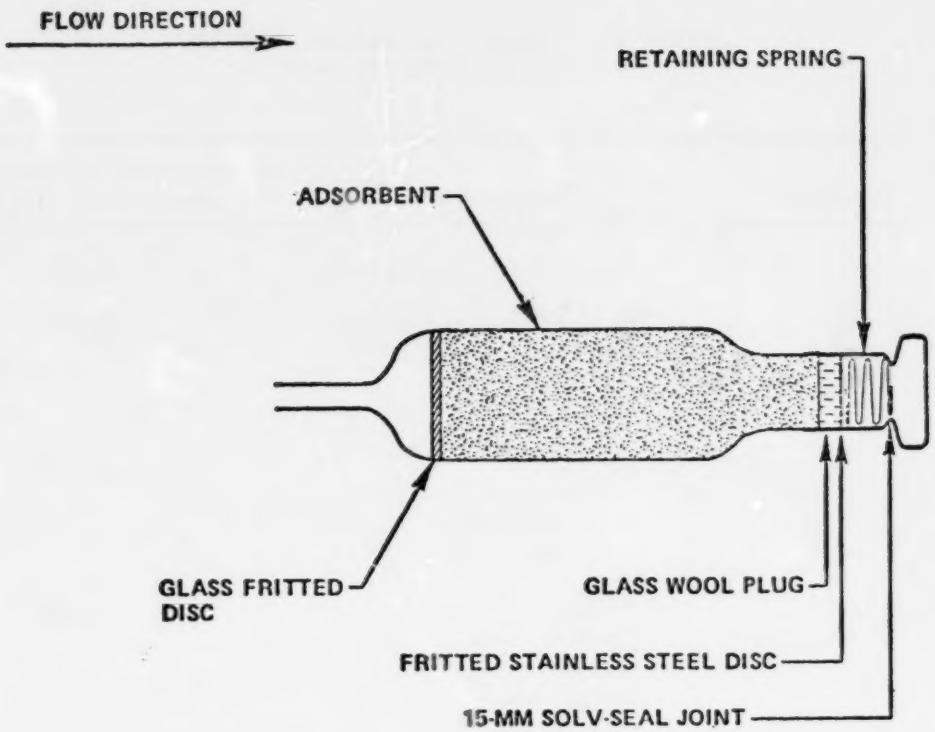



Figure 4. Adsorbent sampling system.

TABLE 9. ORGANIC FRACTIONS MEASURED WITH ADSORBENT SAMPLER

| Fraction | Compound type | Approximate Infrared sensitivity (µg) |
|---|---|---|
| 1 | Aliphatic hydrocarbons | 1.10 |
| 2 | Aromatic hydrocarbons<br>POM<br>PCB<br>Halides | 1.10 |
| 3 | Esters<br>Ethers<br>Nitrocompounds<br>Epoxides | 0.1-1 |
| 4 | Phenols<br>Esters<br>Ketones<br>Aldehydes<br>Phthalates | 0.1-1 |
| 5 | Phenols<br>Alcohols<br>Phthalates<br>Amines | 0.1-1 |
| 6 | Amides<br>Sulfonates<br>Aliphatic acids<br>Carboxylic acid salts | 0.1-1 |
| 7 | Sulfonates<br>Sulfoxides<br>Sulfonic acids | 0.1-1 |
| 8 | Sulfonic acids | 0.1-1 |

a porous polymer absorbent trap. Often these emissions cannot be separated from the forming emissions because a common ventilator is used for both areas. A separate postforming test was performed in those few plants which had separate ventilators for this process step.

The manufacture of primary flat glass products does not involve the application of decorative sprays or lubricity coatings. The major post-forming process step is that of annealing to remove detrimental, residual stresses. In the flat glass industry, this step utilizes natural gas as a fuel, and combustion is expected to be near perfect. Thus, because emissions are expected to be minimal and because no direct gas stream (e.g., stack) is available, no postforming tests were attempted in flat glass plants.

## PRODUCT PACKAGING

Emissions from the product packaging step are expected to be non-existent or negligible. If emissions are present, they are in the form of paper dust from the packaging materials, which is a fugitive emission. Therefore, no testing has been done for this process.

## SECTION 5

## RESULTS AND DISCUSSION

The data obtained from the field sampling are presented by the process subdivisions from which they have been generated, and when appropriate are given both for the glass industry as a whole and by the industry segment. When possible, the field data are further subdivided by glass composition and fuel type. The field data are treated statistically to compare results among various segments of the glass manufacturing industry. The primary statistics are: sample mean ($\bar{X}$), sample variance ($\sigma$), and confidence interval (taken at 10 percent significance) as percent of the mean. Also, regression analyses are used when they are appropriate. They include statistical measures of correlation and confidence of a real relationship (i.e, nonzero slope). The results of these statistical treatments are used to compare the field sampling to the information that has been reported in the SAD's (Table 10).

### BATCHING AND MIXING

As discussed in Section 4, no emissions testing was conducted for batching and mixing. However, observations made by testing personnel while onsite indicated that the dusting from batch preparation and conveying was generally confined within the buildings. All of the plants in which these observations were made were equipped with baghouses, and no visible emission was seen from the exit side of this control device. Therefore the conclusion reached by the SAD's, that only fugitive emissions escape from this process step, appears correct.

### MELTING

The glass melting operation is the largest source of emissions for the glass manufacturing industry, and consequently, most efforts in this research program have been directed toward measuring the emission rates of various criteria pollutants that are produced from this high-temperature process. Two sources of data have been used to gather information for this process step: actual testing under EPA contract, and data voluntarily provided by some participating manufacturers. Most of the discussions in this report will be directed toward the data gathered by onsite testing. These data are presented in Table 11 for the 13 manufacturing operations that voluntarily

TABLE 10. FURNACE EMMISSIONS[a] REPORTED IN THE SAD APPENDICES

| Source | Production Gg/yr | Particulates Mg/yr | $SO_x$ Mg/yr | $NO_x$ Mg/yr | CO Mg/yr | HC Mg/yr |
|---|---|---|---|---|---|---|
| Container Glass | 52.744 (±21.11) | 37.294 (±27.72) | 91.691 (±21.41) | 163.578 (±26.80) | 3.278 (±10.94) | 4.07 (±2.51) |
| Pressed and Blown Glass | 25.05 (±26.48) | 67.520 (±23.52) | 51.52 (b) | 41.071 (±48.61) | 3.000 (b) | 4.0 (b) |
| Flat/float Glass | 162.000 (b) | 131 (b) | 349 (b) | 620 (b) | - - | - - |

[a] Table entries are: $\bar{x}$
(90% confidence interval as percent of $\bar{x}$)

[b] Insufficient data for calculating confidence interval

TABLE 11. SUMMARY OF ONSITE EMISSIONS TESTING DATA

| Data Source | Process Rate, kg/hr (lb/hr) | Fuel | Emissions | | | | |
|---|---|---|---|---|---|---|---|
| | | | Total Particulate, kg/hr (lb/hr) | $SO_x$, kg/hr (lb/hr) | $NO_x$, kg/hr (lb/hr) | CO, ppm | HC, ppm |
| Container | | | | | | | |
| A | 5,470 (12,058) | No. 6 Oil | 8.27 (18.2) | 16.21 (35.7) | 15.74 (34.7) | Trace[a] | Trace[a] |
| D | 8,391 (18,499) | No. 6 Oil | 11.5 (25.3) | 24.3 (53.6) | 22.3 (49.1) | Trace[a] | Trace[a] |
| E | 7,274 (16,039) | No. 2 Oil | 4.40 (9.71) | 11.3 (25.0) | 15.8 (34.9) | 5 | 0 |
| H | 6,227 (13,728) | No. 6 Oil | 6.12 (13.5) | 23.8 (52.3) | 13.6 (29.9) | Trace[a] | Trace[a] |
| I | 5,162 (11,380) | No. 6 Oil | 6.62 (14.6) | 53.35 (117.6) | 12.28 (27.1) | <1 | 56.2 |
| J | 5,384 (11,871) | No. 6 Oil | 15.0 (33.0) | 54.10 (119.3) | 13.49 (29.74) | 47.85 | 6.96 |
| L | 9,873 (21,767) | Gas | 11.98 (26.7) | 25.21 (55.57) | 48.00 (106) | 4.44 | 7.5 |
| M | 11,204 (24,701) | Gas | 9.58 (21.1) | 39.22 (86.46) | 29.50 (65) | 17.1 | 9.15 |
| Pressed and blown glass | | | | | | | |
| C (Soda/lime) | 1,657 (3,653) | Gas | 1.8 (3.9) | 0.46 (1.013) | 8.94 (19.7) | 30 | <5 |
| F (Borosilicate) | 931 (2,053) | Gas | 3.0 (6.6) | Trace[a] | 1.0 (2.3) | Trace[a] | 13 |
| G (Borosilicate) | 950 (2,094) | No. 2 Oil | 3.4 (7.6) | 2.2 (4.8) | 6.0 (13.3) | Trace[a] | 7 |
| K (Lead) | 811 (1,789) | Gas | 3.14 (6.93) | 0.10 (0.21) | 10.0 (22.06) | 2.2 | 9.7 |
| Flat B | 15,876 (35,000) | Gas | 40.0 (88.2) | 68.20 (150.3) | 86.60 (191) | 19.57 | 1.46 |
| $\bar{X}$ | 6,093 (13,434) | | 9.60 (21.12) | 24.49 (53.88) | 21.79 (47.94) | - | - |
| S | 4,521 | | 9.98 | 23.00 | 22.79 | - | - |
| N | 13 | | 13 | 13 | 13 | - | - |
| $\pm$ | 37 | | 52 | 46 | 52 | - | - |

[a] Trace <0.1.

b 90 percent confidence band about the mean as $\pm \bar{x}$.

- Dash indicates insufficient data for calculation.

participated in this testing program. The sources are distributed as follows: eight glass container plants, four pressed and blown glass plants, and one flat glass plant. Data voluntarily supplied by various manufacturers are summarized in Table 12. These voluntarily supplied data are distributed among the three major glass manufacturing segments in the following manner: one glass container furnace, thirteen pressed and blown glass furnaces, and four flat glass furnaces. Specific results and conclusions drawn from the data in Tables 11 and 12 are presented individually for each criteria pollutant sampled.

Oxides of Sulfur

The criteria pollutants with the largest emission rates are the oxides of sulfur, reported as $SO_x$ because of the various gaseous stages in which they can exist. The summaries in Table 11 indicate that $SO_x$ is the pollutant with the largest emission rate in the container industry, and it is slightly more prevalent than $NO_x$ for the entire glass industry as a whole. For the glass container and pressed and blown glass segments, the information in Table 11 can be used to calculate the range bracketing expected annual emission rates. For the glass container industry, this range is 200 to 342 Mg/yr. A similar calculation for the pressed and blown industry data indicates that the emission rate for $SO_x$ ranges from 0 to 13.4 Mg/yr. These data are compared to those of the SAD's and manufacturers in Table 13. Clearly, the $SO_x$ data from the three sources are at variance. The source of these differences cannot be definitively established. However, the non-standard testing procedures that have been used in the data from which the SAD's have been generated can certainly be expected to contribute to the descrepancies. The lack of manufacturer's data also indicates inconsistancy in obtaining $SO_x$ data. Since the current testing program used standardized procedures, its data is expected to be the most reliable.

Plotting of the $SO_x$ emission rate data indicates that a nonlinear relationship exists between the $SO_x$ emission rate, the process rate, and the fuel-use rate (expressed as equivalent kilowatts). A stepwise regression using the logarithms of these three variables indicates that the logarithm of the $SO_x$ emission rate is a function of the logarithm of the ratio of the process rate to the equivalent kilowatt usage. A linear regression analysis involving these two logarithms gave the following statistical results:

1. Correlation -- 93 percent
2. Unexplained variation -- 17 percent
3. Confidence of nonzero slope -- 99.95 percent

The conclusion that can be drawn from this regression analysis, particularly in light of the statistics it yields, is that most of the $SO_x$ emission rate is related to the amount of material being processed through the melting unit and to the rate at which the fuel is used. The high confidence of nonzero slope clearly indicates that the process rate and fuel use rate are the controlling variables for $SO_x$ emissions. The fact that the regression line explains 75 percent of the variation in the data indicates that the difference in the melting units among the three major manufacturing segments are not as large as indicated by references in the SAD's. The unexplained variation

TABLE 12. SUMMARY OF EMISSIONS TESTING DATA SUPPLIED BY GLASS MANUFACTURERS

| Data source | Process rate, kg/hr (lb/hr) | Fuel | Emissions[a] Total particulate, kg/hr (lb/hr) | $SO_x$, kg/hr (lb/hr) | $NO_x$, kg/hr (lb/hr) | CO, ppm | Gaseous F, kg/hr (lb/hr) |
|---|---|---|---|---|---|---|---|
| Container glass: | | | | | | | |
| MAA | 10,932 (24,080) | - | 3.46 (7.63) | 56 ppm | 60.8 (134) | - | - |
| Pressed and Blown glass: | | | | | | | |
| MA (Borosilicate) | 950 (2,094) | Gas | 0.06 (0.13) | - | 1.000 ppm | - | - |
| MB (Soda/lime) | - | Gas + boost | 6.80 (15) | 10.48 (23.1) | - | - | - |
| MC (Soda/lime) | - | No. 2 Oil + boost | 11.98 (26.4) | 52.94 (116.7) | - | - | - |
| MD (Soda/lime) | 8,184 (18,027) | Gas | 2.00 (4.62) | - | <300 ppm | - | - |
| ME (Lead) | 1,678 (3,700) | Gas | 10.16 (20.36) | - | <2000 ppm | - | - |
| MF (Borosilicate) | - | No. 2 Oil | 2.68 (5.9) | 5.49 (12.1) | -[b] | - | - |
| MG (Borosilicate) | 2,703 (5,958) | Gas + boost | 0.84 (1.85) | - | Trace[b] | - | - |
| MH (Fluoride) | 2,326 (5,128) | Gas | 3.12 (6.89) | - | Trace | - | - |
| MI (Fluoride opal) | - | Gas | 12.79 (28.2) | - | - | - | 21.64 (47.7) |
| MJ (Lead) | - | Gas | 6.35 (14) | - | -[b] | - | - |
| ML (Borosilicate) | 3,579 (7,801) | No. 5 Oil + boost | 1.83 (4.03) | - | Trace[b] | - | - |
| MQ (Lead) | - | Gas | 9.27 (20.43) | 2 ppm | 543 ppm | 0.1 ppm | - |
| MY (Lead) | - | Gas | 9.27 (20.43) | 0.05 ppm | 533 ppm | 0.1 ppm | - |
| Flat/Float glass: | | | | | | | |
| MR | - | Gas | 21.55 (47.5) | - | - | - | - |
| MS | - | No. 2 Oil | 16.10 (35.5) | - | - | - | - |
| MW | - | Gas | 14.92 (32.9) | - | - | - | - |
| MZ | - | Gas | 8.71 (19.2) | - | - | - | - |
| Total Industry | | | | | | | |
| $\bar{X}$ | 4,336 (9,567) | | 7.88 (17.34) | 25.68 (5.65) | 547 ppm | 0.1 ppm | 21.64 (47.7) |
| S | 0.955 | | 1.455 | 3.250 | 0.779 | - | - |
| N | 7 | | 18 | 3 | 8 | 2 | 1 |
| $\pm$[c] | 0.701 | | 0.597 | 5.479 | 0.522 | - | - |

a No HC data reported.
b Trace <0.1.
c 90 percent confidence band about the mean as % $\bar{X}$.
- Insufficient data

TABLE 13. COMPARISON OF DATA FOR $SO_x$ EMISSION

| Source[a] | Container glass, Mg/yr | Pressed and blown glass, Mg/yr | Flat/float glass, Mg/yr |
|---|---|---|---|
| SAD's | 72-111 | 51 | 349 |
| Site testing | 200-342 | 0-31 | 597 |

(a) Manufacturer's data was not supplied.

(i.e., 25 percent) can probably be attributed to differences in the rebuilding cycles among the tested tanks, operational differences among the different manufacturers, and raw-material differences that occur because of the different glass compositions present in the data. However, none of these differences seriously detract from the large correlation that exists among glass melting tanks using a variety of fuels and melting a variety of glasses.

Oxides of Nitrogen

The oxides of nitrogen created from the high-temperature combustion can take many forms, such as $NO_2$, $N_2O$, NO, etc. and they are therefore listed in Table 11 as $NO_x$. These oxides of nitrogen comprise the criteria pollutant that has the second largest rate of emission from the glass melting furnaces. Table 11 can be used to determine the annual emission rate for comparison with the SAD's. For the glass container industry, the annual emission rate from the tested furnaces is 133 to 241 Mg/yr. A similar calculation for the data from the pressed and blown industry indicates that the annual emission rate for $NO_x$ is 28 to 86 Mg/yr. Since only one flat glass furnace has been tested under this program, a range cannot be calculated. The comparison of these ranges to the data from the SAD's is shown in Table 14. The agreement between these two data sources indicates that the existing $NO_x$ data is adequate. The lack of manufacturer's data can probably be attributed to a low priority for $NO_x$ testing.

Since $NO_x$ production is the result of a high-temperature combustion process, a regression between the $NO_x$ emission rate and fuel use rate (expressed as equivalent kilowatts) is not unreasonable. The results of such a linear regression are as follows:

1. Correlation -- 98 percent
2. Unexplained variation -- 4 percent
3. Confidence of nonzero slope -- 99.95 percent

The data that have been used to generate this regression line span a large range of glass compositions, through-put rates, and melter sizes. Yet these statistics strongly indicate that a real, positive, linear relationship exists between the $NO_x$ emission range and the fuel use rate (i.e., an increase in fuel use increases the $NO_x$ emission rate). Therefore, in light of the wide range of conditions that have been used to generate this line, the differences among the various industries for $NO_x$ emissions are not large. Moreover, the regression indicates that the rate of $NO_x$ emissions is most strongly dependent on the use of combustion air (proportional to fuel-use rate) and is little influenced by the temperature of the operation. Such a conclusion is not unexpected, because most glass furnaces generally operate in the same high flame temperature realm (above 1400 C). This conclusion is somewhat at variance with the brief $NO_x$ discussions in the SAD's. Those discussions, however, are based on an increase in furnace superstructure temperature which may not necessarily correspond to an increase in flame temperature. Also, the increased superstructure temperature is accomplished by increased flows of fuel and combustion air. Thermodynamic calculations

TABLE 14. COMPARISON OF DATA FOR $NO_x$ EMISSION

| Source[a] | Container Glass, Mg/yr | Pressed and blown glass, Mg/yr | Flat/float glass, Mg/yr |
|---|---|---|---|
| SAD's | 120-208 | 21-41 | 620 |
| Site testing | 133-241 | 28-86 | 759 |

(a) Manufacturer's Data not supplied.

show that at 1700 K (1427 C) the free energies of formation range from -11730 to -16830 cal/mole for $N_2O$ (g), NO (g), or $NO_2$ (g). These free energies are clearly large enough to result in $NO_x$ formation. As larger concentrations of reactants ($N_2$ and $O_2$) are supplied, larger concentrations of $NO_x$ can be expected. The results of the regression analysis are completely consistent with these thermodynamic considerations. Therefore, the cursory treatment in the SAD's is insufficient to adequately explain the controlling variables in $NO_x$ formation.

Total Particulate Matter

Particulate emissions from glass manufacturing furnaces have been routinely reported by the manufacturers, as is evidenced by the data in Table 12. The particulate emission data listed in Table 11 are the total measured--the particulate catch from the probe, the filter, and the impingers. The summaries in Table 11 can be used to calculate the range of total particulate emission that can be expected from the various industries, and these values can be compared to those reported in the SAD's. For the glass container industry, the total particulate emission rate per furnace should range from 65 to 96 Mg/yr. For the pressed and blown glass industry, the total particulate emission rate per furnace should range between 20 and 30 Mg/yr. Again, firm comparison for the flat glass industry cannot be made because only one flat glass furnace has been included in this program. The comparison among the onsite data, the SAD's, and manufacturer's data is shown in Table 15. For both the container glass and flat glass emission, the onsite data from this program is nearly double that in the SAD's and manufacturer's data. This circumstance is the result of reporting only the front-half catch of the particulate train in the SAD and manufacturer's data. The lower value for the onsite data range in the pressed and blown industry may reflect an improved effort toward reducing the particulate emission rate, or it may reflect a production rate for the plants tested in this program that is lower than the industry norm. The data collected are not adequate for statistically establishing the real source of the lowered emission rate in the pressed and blown industry segment.

Several literature references in the SAD's for the glass industry indicate that a relationship may exist between the logarithm of the particulate emission rate and the process rate. The field-test data have been put through a linear regression in those two variables with the following result:

1. Correlation -- 92 percent
2. Unexplained variation -- 17 percent
3. Confidence of a nonzero slope -- 99.95 percent.

The robustness of these statistics for the regression indicate that for a particulate emission rate, the differences among the three manufacturing segments of the glass manufacturing industry are not as large as expected. The positive, nonzero intercept for the regression line indicates that a minimum particulate rate exists when the process rate is zero. That rate is attributable to vapor phase formation of particulates from the

TABLE 15. COMPARISON OF DATA FOR PARTICULATE EMISSION

| Source | Container glass Mg/yr | Pressed and blown glass Mg/yr | Flat/float glass Mg/yr |
|---|---|---|---|
| SAD's | 28-47 | 52-84 | 131 |
| Site testing[(a)] | 65-96 | 20-30 | 350 |
| Manufacturer's Data | 29 | 47-52 | 126 |

(a) Total catch -- front half plus back half

combination of combustion products and species evaporated from the molten bath while the melting unit is in a soaking status.

Particulate emissions from the glass melting furnace are composed of three parts--that which exists at stack temperature, that which is formed on cooling to the filter temperature (120 C), and that which is formed from either cooling to the impinger temperature (ambient), or by reaction with impinger media. Table 16 provides a summary of the fraction of particulates encountered in each of these three portions of the sampling train. Quite clearly, most of the particulates already exist by the time the gas stream has cooled to the stack temperature, and more than 80 percent of the particulate matter has been formed by the time the gas stream has cooled to the filter temperature. These results are used to provide a justification for requiring only the front half of the sample train (probe plus filter) to be considered when reporting particulate emission data. However, the remaining material that condenses in the impingers does account for 15 to 18 percent of the total particulate emission rate, a nontrivial amount of matter.

The chemical composition of the particulate matter captured in the three portions of the sampling train is partially listed in Table 17, which presents the three most prevalent elements (in excess of 1,000 ppm), determined by optical emissions spectroscopy. These data indicate that the captured particulates are a complex mixture of the oxides, or perhaps sulfates, of the constituents that are most easily volatilized from the surface of the molten glass. The presence of aluminum (probably as $Al_2O_3$) indicates that some of the particulate matter may result from carryover of very fine dust in the combustion products, since alumina does not easily volatilize from the molten glass. The high fraction of aluminum in the impingers further indicates that the particulate size for that species is too small to be trapped in either the probe or the filter and must contact the liquid media before it can be removed from the gas stream. This information does not exclude the possibility that some of the species captured in the impingers result from the chemical reaction between a vapor species from the glass melting furnace and the vapor species from other processes in the immediate geographical vicinity of those furnaces. The summaries in Table 17 also indicate that boron, lead, and arsenic are not particularly large contributors in total weight to the emissions from glass melting furnaces. Nevertheless, these species can be particularly harmful in the immediate vicinity of those furnaces which emit them.[7] Petrographic analyses of some electrostatic precipitator dusts that have been sent by the participating manufacturers indicate that all of the particulate matter is quite small. These dusts are primarily from furnaces that produce borosilicate and lead glasses. For the borosilicate dust, most of the particulate is found to be optically anisotropic and is probably either $B_2O_3$ or $H_3BO_3$. The rest of the particulate dust is isotropic and is probably very fine glass. Also, presurvey visits to a plant that melts lead glass have indicated that the stack is lined with a very fine yellow dust that is probably a compound of lead. This observation indicates that lead compounds such PbO and $PbSO_4$ form a significant portion of the particulate matter from furnaces that

TABLE 16. PERCENTAGE OF PARTICULATE LOADING IN THE SAMPLING TRAIN

| Source | Stack | Filter + Probe | Impingers |
|---|---|---|---|
| A | 76.8 | 3.6 | 19.6 |
| B | 17.3 | 24.7 | 58.0 |
| C | 57.6 | 0.0 | 42.4 |
| D | 78.0 | 6.1 | 15.9 |
| E | 40.6 | 52.0 | 7.4 |
| F | 89.4 | 8.3 | 2.3 |
| G | 77.3 | 18.7 | 4.0 |
| H | 80.2 | 10.4 | 9.4 |
| I | 59.8 | 21.6 | 18.6 |
| J | 30.8 | 57.0 | 12.2 |
| K | 58.0 | 33.0 | 9.0 |
| L | 55.7 | 27.0 | 17.3 |
| M | 50.0 | 33.9 | 16.1 |
| $\bar{X}$ | 59.4 | 22.8 | 17.9 |
| S | 21.1 | 17.8 | 15.7 |
| N | 13.0 | 13.0 | 13.0 |
| ± [a] | 13.0 | 29.0 | 33.0 |

a 90% confidence band about the mean as % $\bar{X}$.

TABLE 17. MOST PREVALENT CHEMICAL SPECIES[a] IN PARTICULATE TRAIN[b]

| Source | Probe | | | Filter | | | Impingers | | |
|---|---|---|---|---|---|---|---|---|---|
| | 1st | 2nd | 3rd | 1st | 2nd | 3rd | 1st | 2nd | 3rd |
| A | Si | Na | Al,Ca | Na | Al | K | Al | - | - |
| B | Ca | Na,Si | Mg | K | - | - | Si | - | - |
| C | Ca | Mg | Na | Ca | Na | As | Ca,Si | Na | Mg,Fe |
| D | (c) | (c) | (c) | (c) | (c) | (c) | (c) | (c) | (c) |
| E | Na | K | Ca,Si | Na | K | Ca | Na | Ca,Si | K |
| F | Na | B | K | Na | B,K | As | Si | Ca,Al | K |
| G | Na | B | K | Na | K | B | Ca | Al | Si,Na |
| H | (c) | (c) | (c) | (c) | (c) | (c) | (c) | (c) | (c) |
| I | Na | Ca | K | Na | K | Ca | Sn | - | - |
| J | Na | K | V | Na | K | Ca | Sn | Si | - |
| K | Fe | Pb | K | Pb | K | Na | Ca | Na | Sn |
| L | Na | Ca | K,Cr | Na | K | Ca | Sn | Mg | - |
| M | Na | Ca | Mg | Na | K | Ca | Sn | - | - |

a Determined by optical emission spectroscopy to be above 1,000 ppm.
b EPA Method 5.
c No data available.

melt lead glasses. Unfortunately, this plant could not be scheduled as one of the tested sites, and no quantitative information is available from it.

The size distribution of the particulate matter was determined with an Anderson cascade impactor. Representative plots of particle sizes captured are shown in Figure 5. These graphs indicate that there is a slight difference among the various melting operations, but that all of the particulate matter is very fine, with the median diameter resting somewhere between 0.26 and 0.94 μm. Table 18 provides a summary of the size distribution data that have been gathered from the tested plants. That summary indicates that the median diameter is between 0.5 and 0.83 μm (90-percent confidence band at about the mean value) and that 79 to 88 percent of the collected particulates are less than 3 μm in diameter. In other words, the vast majority of particulate matter emitted from glass melting furnaces is within the respirable size range. No relationship between particle size and stack temperature can be established. Therefore, the size distribution of the particulate matter captured from glass melting furnaces is established by the time the gas stream has cooled to the stack temperature.

### Other Emissions

The remaining criteria pollutants that can be expected from the glass melting operation are unburned hydrocarbons and carbon monoxide that result from incomplete combustion of the fossil fuels. Both Tables 11 and 12 present data for these two species, but data on all potential emissions are not available. Clearly, those instances that do not yield HC and CO are the ones in which combustion is complete. The data in both of these Tables indicate that for both criteria pollutants, trace amounts to amounts that are less than 16 ppm can be generated from the glass melting furnaces and these values agree with those extracted from the SAD's (Table 10).

## FORMING AND POSTFORMING

The forming and postforming emissions can consist of organics, acid fumes, and tin vapors. These emissions are the most difficult to characterize because they are usually released inside the building and escape to the atmosphere through roof-top ventilators. The organic emissions from various participating manufacturers have been characterized using the previously described test procedures. Tin vapors from the production of flat glass, and acid fumes from etching and frosting operations have been surveyed from data sent by manufacturers in those industries.

### Organic Emissions

Organic compounds are released to the atmosphere from sprays and lubricants used in the forming area to effect release of the finished product from the mold, and in the postforming area from lubricity spraying. In general, the emissions from these two operations are not separable because of the construction of the roof-top ventilators. However, the manufacturing facility at one of the tested plants has separate roof-top ventilators over the forming and postforming areas, and separate measurements have been taken

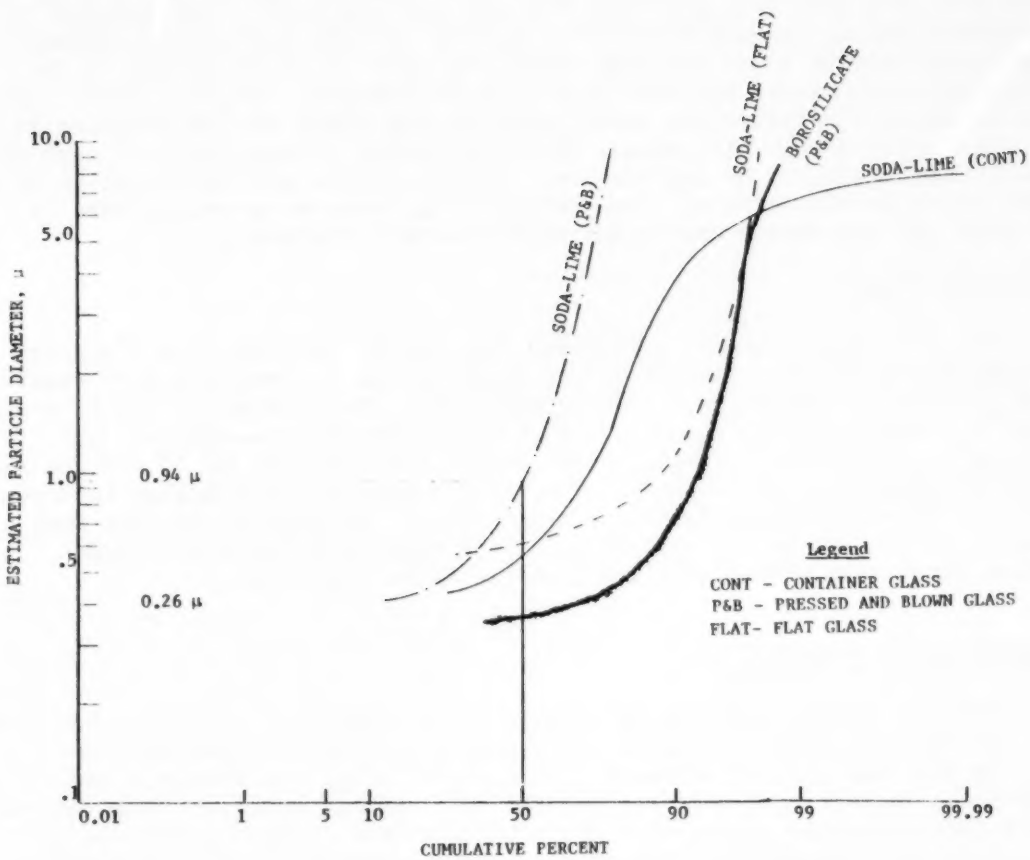



Figure 5. Typical particle size distribution.

TABLE 18. PARTICULATE SIZE DATA

| Source | Median diameter, μ | Respirable[a] fraction, % | Stack temperature, C |
|---|---|---|---|
| A | 0.68 | 80.5 | 158 |
| B | 0.60 | 96.0 | 293 |
| C | 0.90 | 67.0 | 456 |
| D | 0.55 | 87.1 | 218 |
| E | 0.25 | 87.5 | 276 |
| F | 0.36 | 96.0 | 269 |
| G | 0.33 | 98.4 | 257 |
| H | 0.72 | 85.8 | 220 |
| I | 1.20 | 64.0 | 432 |
| J | 1.30 | 64.0 | 449 |
| K | 0.49 | 89.8 | 904 |
| L | 1.04 | 82.0 | 244 |
| M | 0.82 | 83.1 | 249 |
| $\bar{X}$ | 0.71 | 83.2 | 341 |
| S | 0.33 | 11.7 | 194 |
| N | 13.0 | 13.0 | 13 |
| ±[b] | 17.0 | 5.0 | 38 |

a Cumulative fraction < 3μm.
b 90% confidence band as % $\bar{X}$.

at these two locations.

Table 19 lists the total loading and the weight fractions of the eight classes of organic compounds that can be detected with the porous polymer adsorbent trap. Statistics for the loading (expressed as $Mg/nm^3$, or grains/DSCF) show a large variability indicative of the widely-ranging practices in the application of the various release and lubricity compounds. The statistics for the eight organic fractions are also quite variable, but Table 19 does indicate that most of the organics can be found in Fractions 1 through 3 and 7 through 8.

One of the tested manufacturing facilities has separate ventilators over the forming and postforming areas, and this construction allows separate analyses to be conducted in these ventilators. A schematic plan view of that facility is given in Figure 6 and the results of the sampling of this facility are shown in Table 20. As shown in this table, the forming emissions tend to generate the organic materials that are captured as the first three fractions in the porous polymer adsorbent traps. The postforming emissions tend to favor the species that can be captured as the final three fractions of the adsorbent trap. Care must be exercised in interpreting Table 20, because it represents data from only one plant.

Emissions from the forming operation present a particularly difficult control problem because no specific route for the gas stream is provided--it merely exits through a ventilator in the roof. Some glass manufacturers are replacing some of the oils and sprays used for lubrication with water soluble silicons and are thereby reducing the output of organic emissions from this process step. This replacement is slow, however, because of the large financial penalty that can be expected if production is lowered by unforeseen problems. Moreover, much more research is needed before the severity of the emission problem for this process step can be properly established. If that research indicates that a real emission problem does exist, then the most reasonable means of reducing that problem is to find replacements for the oils and sprays currently in use. These replacements can only be established through further research that defines the specific functions and mechanisms involved in the release aid provided by these dopants.

Comparison of data in Table 19 and 20 to those contained in the SAD's is inappropriate because the latter list the data as estimated emission factors or weight of emission per weight of product. The data gathered onsite for organic emissions cannot be converted into such units because the volume flow through the roof-top ventilators is not calculable. However, these onsite data are more reliable than the mass balance estimates in the SAD's.

## Acid Fumes

Acid-fume emissions are generated in the pressed and blown glass industry from etching and polishing, which are postforming operations. Uncontrolled emissions from these processes have not been directly measured in this program. However, some manufacturers did supply data from their own files (Table 21).

TABLE 19. PERCENT BY WEIGHT OF CAPTURED ORGANIC EMISSIONS FROM FORMING AND POSTFORMING OPERATIONS

| Source | Loading mg/m$^3$ [grains/dscf] | Aliphatic hydrocarbons (1) | Aromatic hydrocarbons, POM, PCB, halides, (2) | Esters ethers, nitro compounds, halides (3) | Phenols esters, ketones, aldehydes, phthalates (4) | Phenols, alcohols, phthalates, amines (5) | Amides sulfonates, aliphatic acids, carboxylic, acid salts (6) | Sulfonates, sulfoxides, sulfonic acids (7) | Sulfonic, acids (8) |
|---|---|---|---|---|---|---|---|---|---|
| A (Container) | 7.482 [0.00326] | 28.9 | 2.5 | 10.2 | 26.3 | 8.0 | 3.3 | 18.3 | 2.5 |
| C Pressed and blown | 5.694 [0.00248] | 52.2 | 3.9 | 2.2 | 4.4 | 2.0 | 6.8 | 20.6 | 7.0 |
| I (Container) | 0.979 [0.00426] | 5.9 | 13.7 | 63.4 | 7.5 | 0.0 | 0.0 | 8.8 | 0.6 |
| J (Container) | 0.966 [0.00420] | 11.0 | 67.8 | 0.0 | 0.0 | 2.4 | 0.0 | 18.8 | 0.0 |
| L (Container) | 8.046 [0.0035] | 9.4 | 4.7 | 1.0 | 0.0 | 1.1 | 8.7 | 7.7 | 67.3 |
| M (Container) | 15.852 [0.0069] | 1.4 | 1.8 | 0.9 | 0.1 | 3.6 | 11.0 | 22.2 | 58.8 |
| $\bar{X}$ | 6.503 [0.00283] | 18.1 | 15.7 | 13.0 | 6.4 | 2.8 | 5.0 | 16.1 | 22.7 |
| S | 5.5 [0.00240] | 19.1 | 25.9 | 25.0 | 10.2 | 2.8 | 4.6 | 6.2 | 31.5 |
| ±[a] | 51 (51) | 60.0 | 99.0 | 115.0 | 96.0 | 60.0 | 55.0 | 23.0 | 84.0 |

a 90 percent confidence band around the mean as % $\bar{X}$.

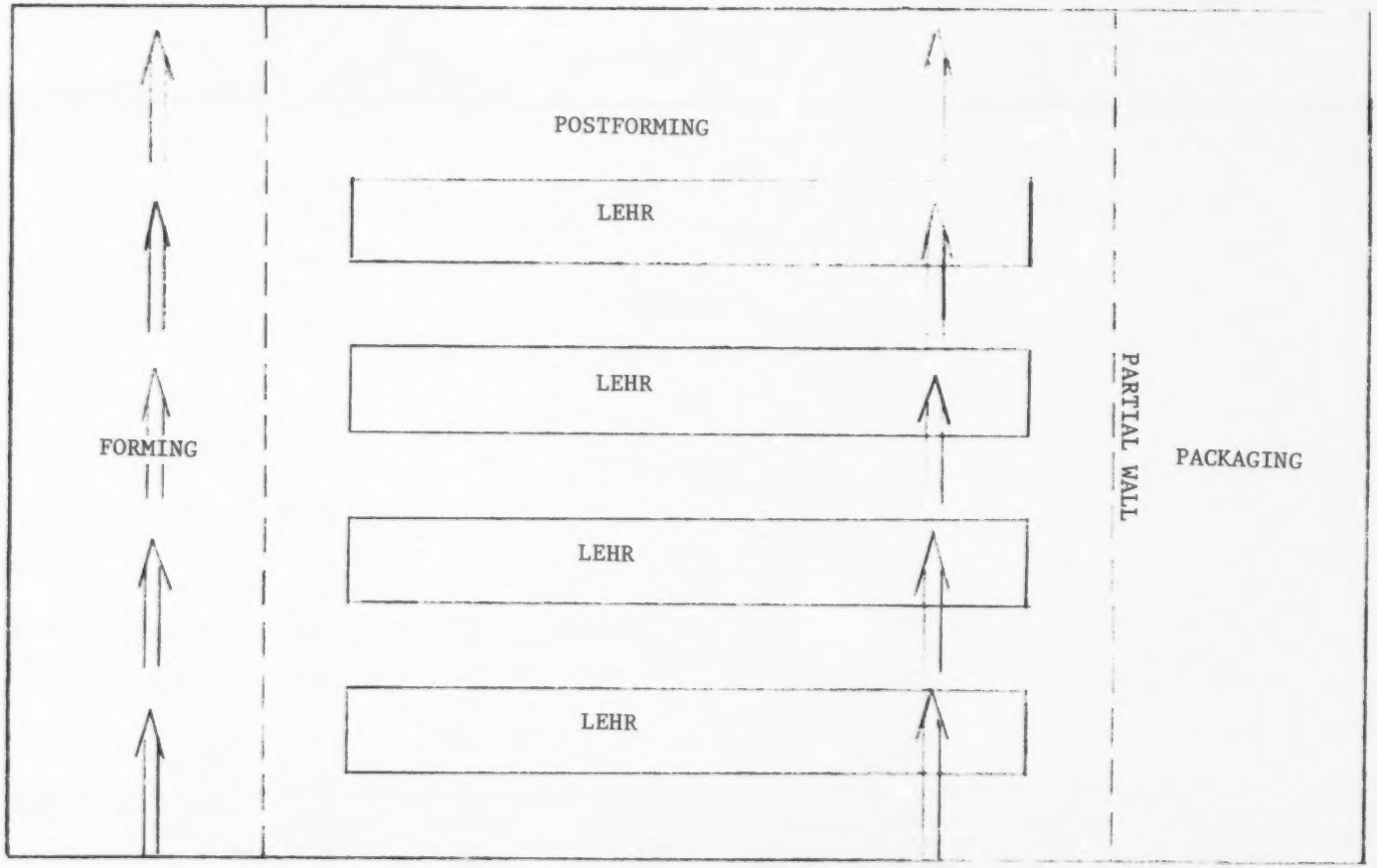



Figure 6. Sampling locations as viewed from the ventilator (arrows indicate sampling locations).

TABLE 20. SEPARABLE FORMING AND POSTFORMING ORGANIC EMISSIONS

| Captured species | Percent by weight captured | | | |
|---|---|---|---|---|
| | Forming | | Postforming | |
| | (L)[a] | (M)[a] | (L)[a] | (M)[a] |
| (1) Aliphatic hydrocarbons | 22.0 | 4.3 | 0.0 | 0.6 |
| (2) Aromatic hydrocarbons POM, PCB, halides | 11.3 | 10.0 | 0.0 | 0.3 |
| (3) Esters, ethers, nitro-compounds | 1.6 | 3.3 | 0.5 | 0.5 |
| (4) Phenols, esters, ketones, aldehydes, phthalates | 0.0 | 0.2 | 0.0 | 0.0 |
| (5) Phenols, alcohols, phthalates, amines | 0.0 | 2.2 | 1.9 | 4.2 |
| (6) Amides, sulfonate, aliphatic acids, carboxylic acid salts | 15.6 | 3.6 | 0.0 | 9.0 |
| (7) Sulfonates, sulfoxides, sulfonic acids | 2.7 | 76.2 | 0.0 | 4.8 |
| (8) Sulfonic acids | 46.7 | 0.0 | 97.6 | 80.6 |
| Loading $\frac{mg}{m^3}$ | 3.035 | 2.157 | 1.962 | 9.848 |
| ($\frac{grains}{dscf}$) | (0.00132) | (0.000939) | (0.000854) | (0.00249) |

a Manufacturer's code

TABLE 21. UNCONTROLLED FUMES FROM ACID ETCHING

| Source | Type of glass | HF , kg/hr | (lb/hr) |
|---|---|---|---|
| MQ | TV | 0.77 | (1.702) |
| $MB_1$ | Soda-lime | 3.6 | (7.9) |
| $MB_2$ | Soda-lime | 3.1 | (6.8) |
| $MB_3$ | Soda-lime | 1.8 | (4.0) |

Since acid-etching procedures have been used in the industry for several years, these values are taken to be representative of the practices in the industry. These values are within the range of information reported in the SAD's (0.18 to 0.96 g of emission/kg of glass production).

Tin Vapors

The flat glass industry manufactures most of its products through the float process. In this process, the molten glass passes over a bath of molten tin to form the final product. This molten tin bath presents a potential source of emissions from the flat glass manufacturing operations. One of the participating manufacturers has supplied data that indicate that the tin usage is approximately 50 lb/yr, or 0.2 mg of tin (vaporized)/kg of glass produced. That value is nearly a factor of ten less severe than the value of 1 mg/kg of glass, as has been reported in the SAD for the flat glass industry.[2] The actual emission factor for tin vapors is probably considerably less than the estimated value of 0.2 mg/kg of glass because most of the tin is lost through adhesion of a very thin tin film to the glass as it is being drawn over the molten tin bath.

Other Emissions

Other potential emissions from the forming and postforming operations include the products of combustion from the annealing layers, organic emissions from the decorating media, and hydrochloric acid vapors from surface treatment operations. The character of these emissions have not been discussed because they either are not present in the tested plants or they are not separable by the techniques used for this study. The largest potential source for these other emissions is the combustion products from the annealing operations. These operations generally use natural gas, and the precise temperature control requires a good control over the combustion process. Such carefully controlled combustion processes have a very low potential for emitting unburned hydrocarbons or carbon monoxide. Therefore, the estimates provided in the SAD's are not expected to be seriously in error with actual practice.

None of the tested manufacturing operations produced articles that were decorated, so no measure of emissions from this type of operation was made. Some of the tested plants did apply lubricity coatings that could potentially yield organic emissions and hydrochloric acid vapors. If organic emissions were present, they were measured with the porous polymer adsorbent traps. On the other hand, hydrochloric acid emissions, if present, were not measurable by the techniques used in this study. Thus, SAD estimates for the emission rates of organics from product decorating and for hydrochloric acid vapors from lubricity spraying operations were not altered as a result of the present study.

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